

reflection (or other higher order peaks) suggests the (100) family of planes of the hexagonal array are parallel to the substrate surface. The width of the (100) reflection does not change significantly with calcination, indicating the good stability of the mesoporous structure. Because of the difficulty in locating the sample in the same position in the X-ray diffractometer before and after calcination, no interpretation can be made on the change in the peak height (i.e., the decrease of the peak height in FIG. 20 does not necessarily indicate a loss of structural order. The d_{100} -spacings are 37.5 Å before and 34.5 Å after calcination for the film prepared with formulation #56483-2; and 36.5 and 33 Å, respectively, for calcination for the film prepared with formulation #56483-5.

The top surface of the spin-coater chuck, on which the cover slips sits during spin coating, consists of a series of radial grooves approximately 1 mm apart. This pattern of grooves also appears in the interference colors of the films spun from both precursor solutions. It is believed that the chuck acts as a heat sink for the thin cover slip. As the solvent evaporates during spin coating, spatial variations in heat transfer through the cover slip results in temperature variations which in turn results in variations in the film thickness.

Closure

While a preferred embodiment of the present invention has been shown and described, it will be apparent to those skilled in the art that many changes and modifications may be made without departing from the invention in its broader aspects. The appended claims are therefore intended to cover all such changes and modifications as fall within the true spirit and scope of the invention.

We claim:

1. A method of making mesoporous silica materials, comprising the steps of

- (a) combining a silica precursor with an aqueous solvent, an acid and a surfactant having an ammonium cation into a silica precursor solution.
- (b) templating the silica precursor with the surfactant and obtaining the mesoporous material from the templated silica precursor.
- (c) forming said silica precursor solution into a preform; and
- (d) rapidly evaporating said aqueous solvent from said preform for obtaining the mesoporous material, wherein the improvement comprises:
 - (i) providing said aqueous solvent in an amount resulting in complete hydrolysis and providing said acid in an amount maintaining a hydrolyzed precursor and avoiding gelation or precipitation; and
 - (ii) providing said surfactant and said silica precursor in a mole ratio that is above a lower mole ratio that produces a non-porous silica phase and below an upper mole ratio that produces a lamellar phase.

2. The method as recited in claim 1, wherein said lower mole ratio is about 0.05.

3. The method as recited in claim 1, wherein said upper mole ratio is about 0.3.

4. The method as recited in claim 1, wherein said acid is added in an amount resulting in a pH of said silica precursor solution of from about 1 to about 4.

5. The method as recited in claim 4, wherein said pH is about 2.

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6. The method as recited in claim 1, wherein the step of forming includes diluting with an alcohol.

7. The method as recited in claim 6, wherein said alcohol is ethanol.

8. The method as recited in claim 1, wherein said aqueous solvent, said acid, and said surfactant are premixed before combining with said silica precursor.

9. The method as recited in claim 1, wherein said mesoporous material is in a geometric form selected from the group consisting of fiber, powder, and film.

10. The method as recited in claim 1, wherein said forming is spin-casting.

11. The method as recited in claim 1, wherein said forming is spraying.

12. The method as recited in claim 1, further comprising adding a pre-polymer or a polymer to said silica precursor solution making a pituitous mixture.

13. The method as recited in claim 1, wherein said forming is drawing.

14. The method as recited in claim 1, wherein said forming is squeeze-rolling.

15. The method as recited in claim 1, further comprising the step of adding a metal compound to the silica precursor solution.

16. The method as recited in claim 15, wherein said metal compound is selected from the group consisting of metal halide, metal nitrate, and combinations thereof.

17. The method as recited in claim 16, wherein said metal halide is a metal chloride.

18. The method as recited in claim 16, wherein said metal is selected from the group of aluminum, iron and combinations thereof.

19. The method as recited in claim 1, wherein said silica precursor is an alkoxide silica precursor or a tetrachlorosilane.

20. The method as recited in claim 1, wherein said aqueous solvent amount is characterized by a ratio of said aqueous solvent to said silica precursor of about 7.

21. The method as recited in claim 1, wherein said acid amount is characterized by a ratio of said acid to said silica precursor of about 0.1.

22. The method as recited in claim 1, further comprising adding a swelling agent to the silica precursor solution.

23. The method as recited in claim 22, wherein said swelling agent is 1,3,5-trimethylbenzene.

24. The method as recited in claim 1, further comprising the step of calcining the mesoporous material.

25. A method of making a mesoporous silica film, comprising the steps of

(a) combining a silica precursor with an aqueous solvent, an acid and a surfactant having an ammonium cation into a silica precursor solution.

(b) templating the silica precursor with the surfactant and obtaining the mesoporous material from the templated silica precursor.

(c) forming said silica precursor solution into a preform; and

(d) rapidly evaporating said aqueous solvent from said preform for obtaining the mesoporous material, wherein the improvement comprises:

(i) said silica precursor is tetraethoxysilane;

(ii) providing said aqueous solvent in a superstoichiometric amount and providing said acid in an amount

maintaining a hydrolyzed precursor and avoiding gelation or precipitation;

(iii) providing said surfactant and said silica precursor in a mole ratio that is above a lower mole ratio that produces a non-porous silica phase and below an upper mole ratio that produces a lamellar phase; and

(iv) said forming includes diluting with an alcohol.

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26. The method as recited in claim 26, further comprising adding a pre-polymer or a polymer to said silica precursor solution making a pituitous mixture.

27. The method as recited in claim 26, wherein said rapidly evaporating is by spin-casting.

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28. A mesoporous silica powder having a plurality of particles comprising:
said plurality of particles in a size range are hollow spheres with mesopores therein.
29. The particles as recited in claim 28, having a diameter of at least 1 micron.
30. A mesoporous silica product, comprising:
a fiber having a diameter of at least 5 microns with mesopores therein,
whereby the mesoporous silica product is formed by a process including any one or
more of the steps including dry spinning, fiber drawing and evaporation.
31. A calcined mesoporous silica film on a substrate formed by a process
comprising any one or more of the steps including spin coating, dip coating and evaporation.
32. A process to form mesostructured films, comprising:
(a) preparing a precursor sol containing a soluble source of a metal oxide, water,
an organic solvent, surfactant, and acid or base catalyst, wherein the surfactant concentration
 c_0 is much less than the critical micelle concentration and the surfactant is present as free
surfactant; and
(b) depositing the precursor sol on a substrate wherein evaporation of solvent and
water causes the formation of said mesostructured films on the substrate surface wherein said
mesostructured films are identified by XRD peaks in the range 2θ - 2° - 6° hexagonal, cubic, or
lamellar electron diffraction patterns.
33. The process of claim 32, wherein said film is used as a membrane, sensor,
catalyst, low dielectric constant interlayer, or optical coating.
34. The process of claim 32, wherein said metal oxide is an oxide of silicon,
aluminum, or combinations thereof.
35. The process of claim 32, wherein the sol is aged prior to film deposition to
affect a change of the film microstructure.
36. The process of claim 32, wherein the sol is aged between 0 to approximately 4
hours prior to film deposition.

37. The process of claim 32, wherein the surfactant includes a cationic surfactant.

38. The process of claim 32 further comprising the step of calcining said film at approximately 400 °C. *degrees?*

39. The process of claim 32, wherein the precursor sol is deposited on a substrate by spin coating. *no support for this temp*

40. A method of making a mesoporous film on a substrate, the method comprising the steps of:

(a) combining a silica precursor with an aqueous solvent, a catalyst and a surfactant into a precursor solution;

(b) depositing said precursor solution in the form of a film onto the substrate; and

(c) removing said aqueous solvent, said catalyst and said surfactant from the film to form a mesoporous film, said removing being performed by any one or more of the steps including spin-coating, rapid evaporation and calcination. *which are they? how done*

41. The method of claim 40 wherein the precursor solution is a silica precursor solution and wherein the surfactant and the silica precursor solution are in a mole ratio that is above a lower mole ratio that produces a non-mesoporous silica phase and below an upper mole ratio that produces a lamellar phase.

✓ 42. A method of making a mesoporous film on a substrate comprising the steps of:

(a) providing a substantially planar substrate;

(b) depositing a precursor solution onto the substrate, the precursor solution including an aqueous solvent, a catalyst and a surfactant;

(c) first removing substantially all of the aqueous solvent to form a templated mesostructured thin film on the substrate; and

(d) second removing substantially all of the catalyst and surfactant to form a mesoporous thin film on the substrate.

43. The method of claim 42 wherein said first removing includes the step of rapidly evaporating by spin coating.

44. The method of claim 42 wherein said second removing includes the step of calcining.

45. The method of claim 42 wherein the aqueous solvent and the catalyst are provided in amounts that maintain a hydrolyzed precursor solution while avoiding gelation or precipitation.

46. The method of claim 42 wherein the precursor solution is a silica precursor solution and wherein the surfactant and the silica precursor solution are in a mole ratio that is above a lower mole ratio that produces a non-mesoporous silica phase and below an upper mole ratio that produces a lamellar phase. *which is*

47. The method of claim 46 wherein the aqueous solvent and the catalyst are provided in amounts that maintain a hydrolyzed precursor solution while avoiding gelation or precipitation.

48. The method of claim 42 wherein the aqueous solvent is provided in a superstoichiometric amount and wherein the catalyst is an acid and wherein the aqueous solvent and the catalyst are provided in amounts that maintain a hydrolyzed precursor solution while avoiding gelation or precipitation.

Sys 49. The method of claim 42 wherein the precursor solution comprises tetraethoxysilane to form a silica thin film on the substrate.

50. The method of claim 49 wherein the surfactant and the precursor solution are in a mole ratio that is above a lower mole ratio that produces a non-mesoporous silica phase and below an upper mole ratio that produces a lamellar phase.

51. The method of claim 50 wherein said first removing includes the step of rapidly evaporating by spin coating and wherein the silica precursor is diluted with an alcohol prior to said spin coating.

52. The method of claim 42 wherein the mesoporous film has a refractive index of less than approximately 1.25.

53. The method of claim 42 wherein the mesoporous film has a refractive index of less than approximately 1.16.

54. The method of claim 42 wherein the mesoporous film has sufficient porosity to result in a low-k dielectric constant of less than approximately 3.0.

55. The method of claim 54 wherein the substrate is a silicon substrate and wherein the mesoporous film is a low-k dielectric film.

56. A method of making mesoporous silica materials, comprising the steps of:

- (a) combining a silica precursor with an aqueous solvent, an acid and a surfactant into a silica precursor solution;
- (b) templating the silica precursor with the surfactant and obtaining the mesoporous material from the templated silica precursor;
- (c) forming said silica precursor solution into a preform; and
- (d) rapidly evaporating said aqueous solvent from said preform for obtaining the mesoporous material, wherein the improvement comprises:
 - (i) providing said aqueous solvent in an amount resulting in complete hydrolysis and providing said acid in an amount maintaining a hydrolyzed precursor and avoiding gelation or precipitation; and
 - (ii) providing said surfactant and said silica precursor in a mole ratio that is above a lower mole ratio that produces a non-porous silica phase and below an upper mole ratio that produces a lamellar phase.

57. A method of making a mesoporous silica film, comprising the steps of:

- (a) combining a silica precursor with an aqueous solvent, an acid and a surfactant into a silica precursor solution;
- (b) templating the silica precursor with the surfactant and obtaining the mesoporous material from the templated silica precursor;
- (c) forming said silica precursor solution into a preform;

- (d) rapidly evaporating said aqueous solvent from said preform for obtaining the mesoporous material, wherein the improvement comprises:
- (i) said silica precursor is tetraethoxysilane;
 - (ii) providing said aqueous solvent in a superstoichiometric amount and providing said acid in an amount maintaining a hydrolyzed precursor and avoiding gelation or precipitation;
 - (iii) providing said surfactant and said silica precursor in a mole ratio that is above a lower mole ratio that produces a non-porous silica phase and below an upper mole ratio that produces a lamellar phase; and
 - (iv) said forming includes diluting with an alcohol.

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Sub 58 58. A process to form mesostructured films, comprising:

(a) preparing a precursor sol containing a soluble source of a metal oxide, water, an organic solvent, surfactant, and acid or base catalyst, wherein the surfactant concentration c_0 is much less than the critical micelle concentration and the surfactant is present as free surfactant; and *112 lig time*

(b) depositing the precursor sol on a substrate wherein evaporation of solvent and water causes the formation of said mesostructured films on the substrate surface.

59. The process of claim 58 wherein the metal oxide, water and organic solvent form an aqueous solvent and wherein the aqueous solvent and the catalyst are provided in amounts that maintain a hydrolyzed precursor sol while avoiding gelation or precipitation.

60. The process of claim 58 wherein the precursor sol is a silica precursor sol and wherein the surfactant and the silica precursor sol are in a mole ratio that is above a lower mole ratio that produces a non-porous silica phase and below an upper mole ratio that produces a lamellar phase.

61. The process of claim 58 wherein the metal oxide, water and organic solvent form an aqueous solvent and wherein the aqueous solvent and the catalyst are provided in amounts that maintain a hydrolyzed precursor sol while avoiding gelation or precipitation.

Sub 62 62. The process of claim 58 wherein the metal oxide, water and organic solvent form an aqueous solvent provided in a superstoichiometric amount and wherein the aqueous

solvent and the catalyst are provided in amounts that maintain a hydrolyzed precursor sol while avoiding gelation or precipitation.

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